Neutron-Induced Hydrogen and Helium Production from Threshold to 100 MeV

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ABSTRACT -- *We are measuring the production cross sections for hydrogen and helium production by neutrons from threshold to 100 MeV on structural materials of importance to the Advanced Fuel Cycle Initiative. These gases are known to be important contributors to changes in the properties of materials subjected to high fluences of neutrons. Our approach is to measure the isotopes of hydrogen and helium as they are emitted in the nuclear reactions such as (n,xp), (n,xd), and (n,xalpha). The data include not only the production cross sections but also the angular distributions and energy spectra of the emitted particles. These extra data test nuclear reaction model calculations, which are used to provide data in evaluated libraries. Both statistical as well as precompound nuclear reaction mechanisms have been identified. Preliminary data have been taken on thin foils of* ^{58}Ni *and* ^{60}Ni *and several thicknesses of natural iron. The results show that the production cross sections generally increase with incident neutron energy. Helium production data on the nickel isotopes is significantly smaller than that given in the ENDF/B-VI evaluated library in the 7-12 MeV neutron energy region. These data will offer reliable source terms for gas production in the assessment of radiation damage to structural materials*

I. INTRODUCTION

Hydrogen and helium are produced when energetic neutrons interact with materials, and these gases can lead to significant changes in materials properties such as embrittlement and swelling. Such effects have already been seen in fission reactors, and new designs from the Advanced Fuel Cycle Initiative with projected higher neutron fluences on structural material are certain to increase interest in these effects. At higher average neutron energies, significant effects are expected in the development of fusion reactors and advanced accelerator applications because of the gas production rates per neutron (proportional to the production cross sections) are higher at these energies. For all of these systems, new materials are being developed with elements for which the gas production cross sections are not well known and the nuclear model predictions have significant uncertainties. Thus there is a need for more experimental data on gas production cross sections to improve the evaluated data libraries and to validate nuclear reaction model codes for calculating the production for elements and isotopes that are difficult to study in the laboratory.

We have begun a program to measure these cross sections on elemental constituents of structural materials. This work is a continuation of measurements made up to 60 MeV on other materials such as silicon[1] and cobalt.[2]

II. EXPERIMENTAL METHOD

Our method is more detailed than simply the measurement of gases produced in a material. Instead we detect protons, alpha particles and other isotopes of hydrogen and helium emitted in reactions induced by neutrons, and we measure their energy and angular distributions. There are several reasons for this approach. First, the measurement of hydrogen and helium after irradiation of materials with monoenergetic neutrons requires a very intense neutron source and has been done only at a few energies, principally at 10 and 14 MeV for helium.[3,4] Hydrogen production is even more difficult to measure directly because of the omnipresence of hydrogen-containing compounds. Thus the measurement of light charged particles is usually the only way to determine gas production cross sections. Second, the additional information from the energy and angular distributions of the emitted light charged particles tests reaction models that are used to calculate gas production more generally for evaluated data libraries used in applications. Third, with a continuous-in-energy, pulsed, "white" source of neutrons, the gas production can be measured as a function of incident neutron energy only if the reaction products are detected with good time resolution of a few nanoseconds. Light charged particles can be so detected.

The present measurements were made at the WNR/LANSCE spallation neutron source[5] on two different flight paths. The WNR/LANSCE neutron source is a pulsed source based on short (sub 1 ns) bunches of 800 MeV protons that are incident on a tungsten neutron-production target. The resulting neutrons are collimated into neutron beams, which then pass along flight paths to the experimental stations. For these measurements, we used flight paths at 90-degrees and 30-degrees with respect to the incident proton beam. The flight path lengths were 9.1 and 15.1 meters respectively. The 30-degree beam line gives relative more neutrons at high energies (see Figure 1) and was chosen to extend the 90-degree measurements to higher neutron energies. The beam was collimated to a 5 cm x 5 cm square for the 90-degree measurements and a 2.2 cm diameter for the 30-degree measurements. The smaller diameter allows the use of samples of smaller area typical of isotopically enriched materials. Time-of-flight techniques are used to deduce the energy of the neutron that induced the reaction.

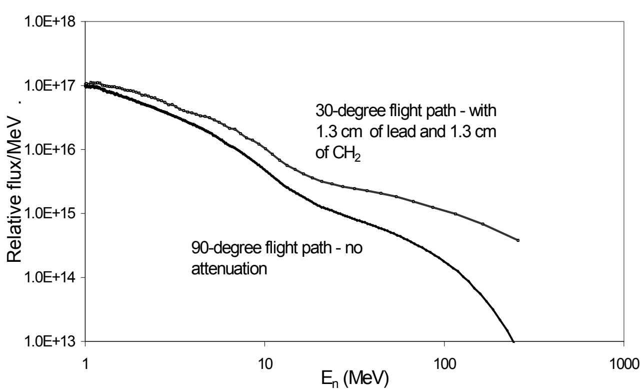

Fig. 1. Shapes of the neutron flux spectra for the 30- and 90-degree flight paths.

An important feature of our experiments is that the data are obtained for the full range of incident neutron energies simultaneously. The test chamber is an evacuated chamber with inside diameter of 55.9 cm to accommodate the test materials and the detectors. The chamber and the detector layout are shown in Fig. 2.

Detector systems for the emitted charged particles are deployed at 4 angles with respect to the incident neutron beam direction. The angles are chosen to cover much of the full angular range so that the angle-integrated production cross section can be reliably determined from the differential data. The detectors are coincidence counters consisting of two Delta-E transmission counters and a stopping E-counter. The first Delta-E counter is either a low pressure proportional chamber filled with xenon gas to a pressure of nominally 3.3 kPa (25 torr) (for relatively highly ionizing particles such as alpha particles) or a thin silicon surface barrier detector (for lower ionizing particles such as protons). The second Delta-E detector is a 500 micron silicon surface barrier detector, which does in fact stop protons below about 12 MeV and alphas below 33 MeV. We use CsI(Tl) detectors with thicknesses of 1 cm and more recently 3 cm for the E-detector to stop more energetic particles. Signals from these detectors undergo preliminary processing near the chamber, and then the processed signals are transported to further electronics and a data acquisition system in a nearby building.

For many of the measurements, the target-out background is small and corrections are not difficult. At the higher neutron energies, however, the collimation is challenged, especially in measurements of proton-production. Protons have longer ranges in materials and consequently the cleanup shielding can behave as a thick sample for the neutrons that penetrate to surfaces viewed by the detectors. At the newer 30-degree station, better collimation and shielding were possible as it was desired to extend the measurements to incident neutron energies up to 100 MeV. An example of the data obtained for an iron sample is given in Fig. 3.

III. EXPERIMENTAL RESULTS

To date preliminary results have been obtained for several thicknesses of iron, one thick chromium sample, and two isotopically enriched samples of nickel, ^{58}Ni and ^{60}Ni. Because the data sets are not fully analyzed at the time this manuscript was due, we present here fully analyzed data for neutron-induced helium production for the nickel isotopes. Some of these data have been presented previously.[6]

These nickel samples were rolled foils 3.38 and 2.98 mg/cm^2 thick for ^{58}Ni and ^{60}Ni respectively. The 90-degree flight path was used and the collimation was larger than in the later design shown above, approximately 5 x 5 cm^2. The emitted alpha particles were detected at angles of 30, 60, 90 and 135 degrees, and the angle and energy distributions of the double differential cross sections were integrated to obtain the total helium production.

The results are shown in Figs. 4 and 5, where they are compared with the ENDF/B-VI evaluations up to 20 MeV and with the LA150 evaluation[7] from 20 to 100 MeV. Comparison with literature values shows reasonable agreement for incident neutron energies below 15 MeV.[6] There are no measurements above 15 MeV in the literature to compare with the present results.

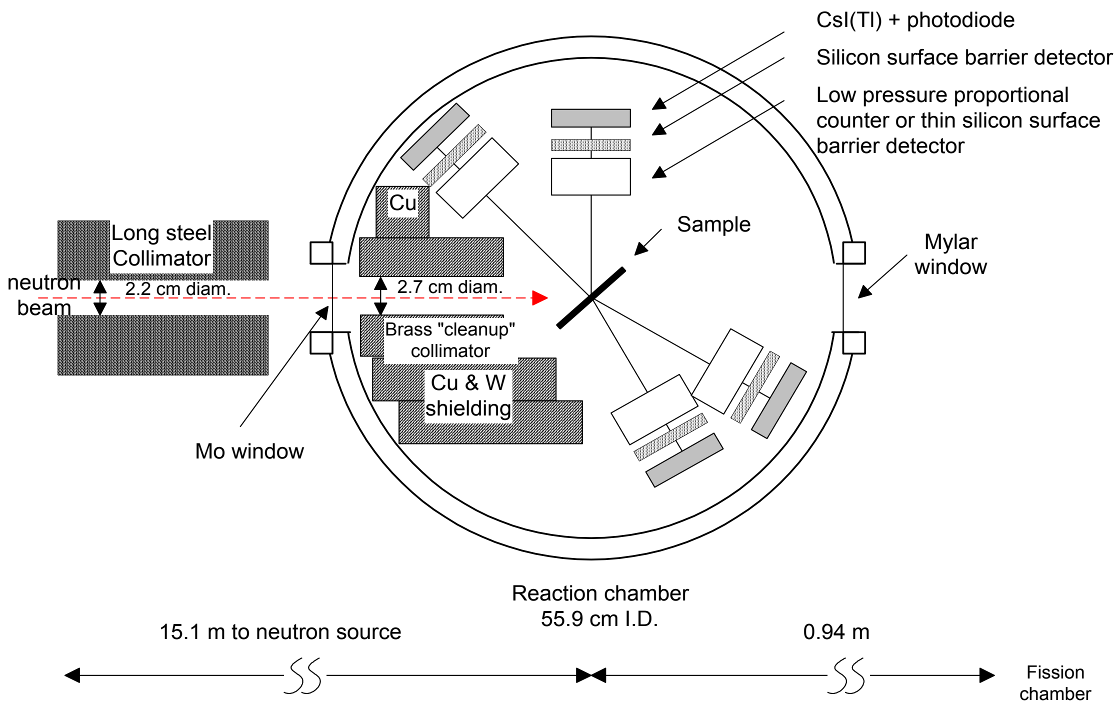

Fig. 2. Layout of apparatus for measuring neutron-induced hydrogen and helium production showing collimation for the neutron beam, target position, array of detectors, and location of fission chamber downstream for measuring the neutron energy-dependent fluence.

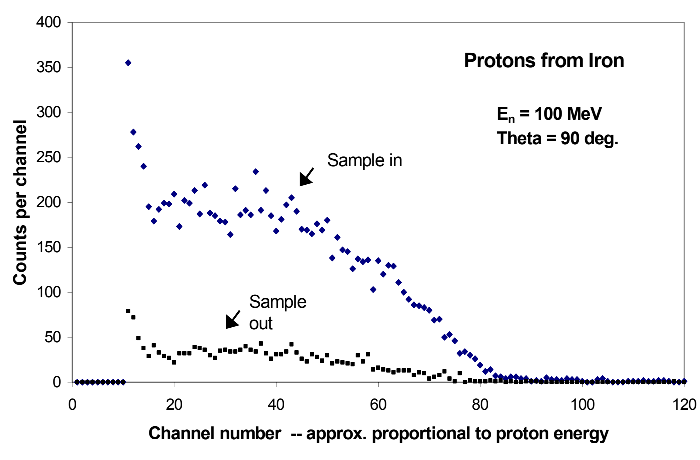

Fig. 3. Sample-in and sample-out measurements of proton production from an iron sample at a neutron energy of 100 MeV. The signal-to-background ratio is good at this neutron energy and significantly better at lower incident neutron energies.

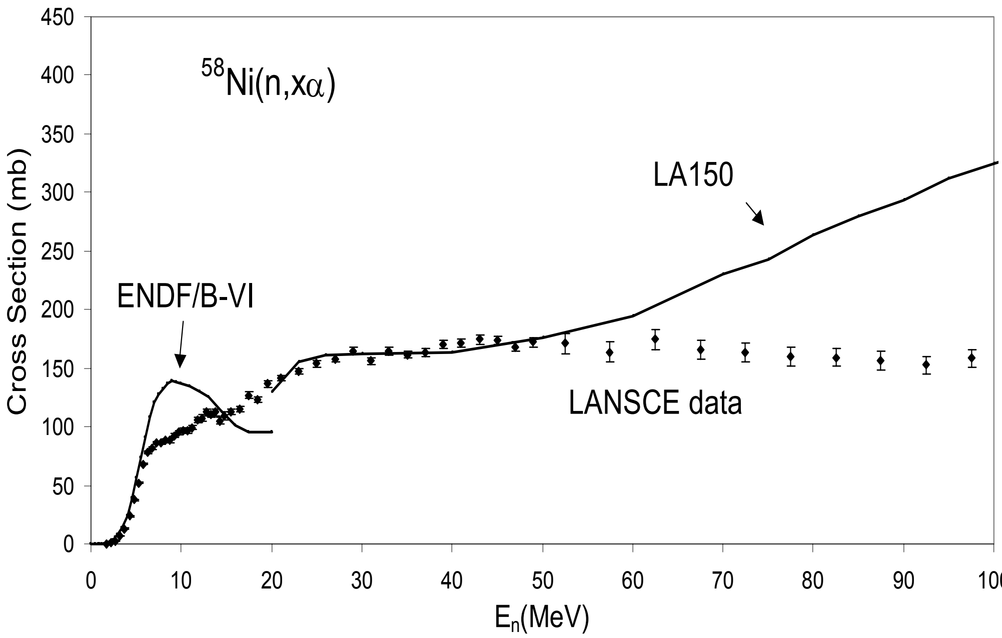

Fig. 4. Cross section for production of alpha particles by neutrons from threshold to 100 MeV on ^{58}Ni. The data up to 50 MeV were presented earlier.[6] The data from 50 to 100 MeV are new. Curves show evaluated data in the ENDF/B-VI and LA150 libraries. The ENDF/B-VI evaluation was made before any of these experimental data were available. The LA150 evaluation took account of the experimental results up to 50 MeV.

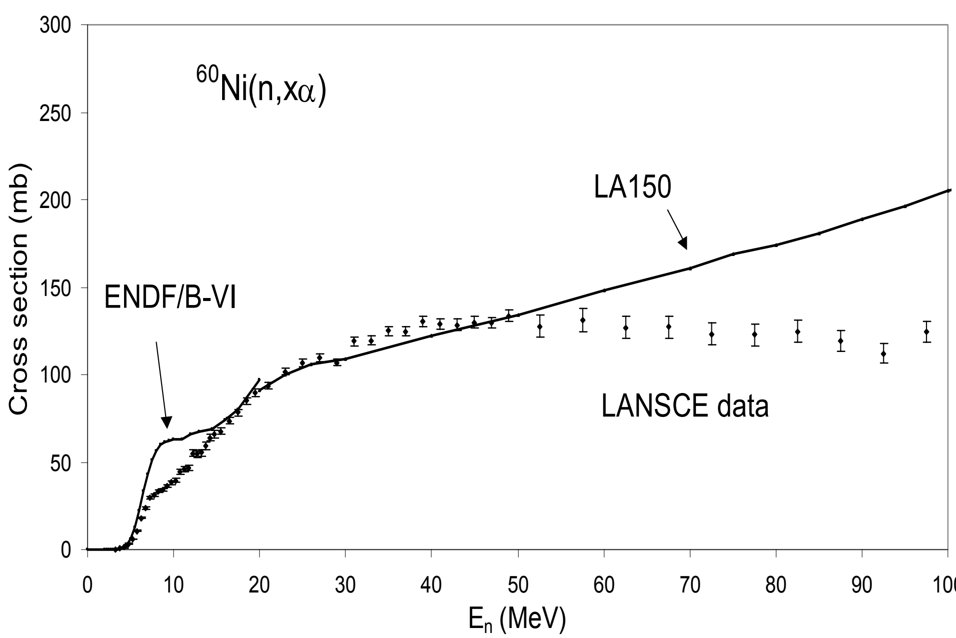

Fig. 5. Cross section for production of alpha particles by neutrons from threshold to 100 MeV on ^{60}Ni. The data up to 50 MeV were presented earlier.[6] The data from 50 to 100 MeV are new. Curves show evaluated data in the ENDF/B-VI and LA150 libraries. The ENDF/B-VI evaluation was made before any of these experimental data were available. The LA150 evaluation took account of the experimental results up to 50 MeV.

It is interesting to compare the charged particle emission spectra for different incident neutron energies. A sample of the alpha-particle emission data for ^{60}Ni is shown in Figure 6. As one might expect, the end point of the emission spectrum increases as the incident neutron energy increases. A further feature to be noted is that there is an increase in the emission cross section for alpha particles above 10 MeV relative to lower energy alphas as the incident neutron energy increases. The shapes of the emitted alpha spectra are indicative of pre-compound particle emission. Nuclear model calculations are underway to try to reproduce these results.

IV. Summary

An experimental facility has been established at the LANSCE/WNR spallation neutron source for measuring hydrogen and helium production cross sections from threshold to 100 MeV. First results show good signal-to-background ratios even at the upper end of this neutron range. Heliium production on nickel isotopes differs in the 8-13 MeV range from the ENDF/B-VI evaluated data libraries for ^{58}Ni and ^{60}Ni and in the 15-20 MeV range for ^{58}Ni. The experimental results above 20 MeV are new and will guide future evaluations and comparisons with nuclear reaction model calculations.

ACKNOWLEDGMENTS

This work has benefited from the use of the Los Alamos Neutron Science Center at the Los Alamos National Laboratory. This facility is funded by the US Department of Energy and operated by the University of California under Contract W-7405-ENG-36.

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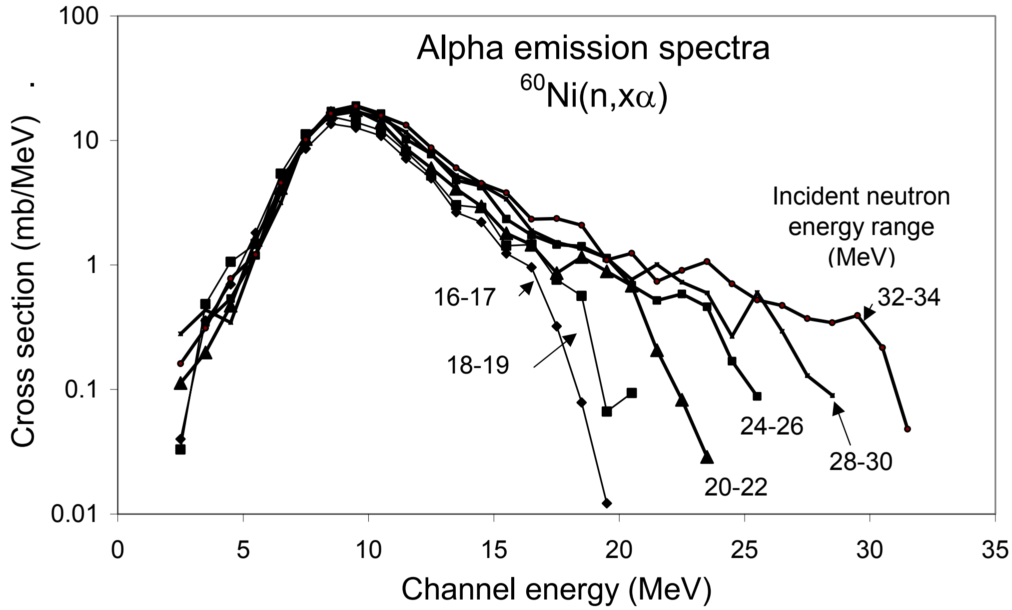

Fig. 6. Emission spectra of alpha particles from bombardment of ^{60}Ni with neutrons of energies indicated in the figure. The common feature is the evaporation peak centered at slightly less than 10 MeV channel energy, which is the sum of alpha particle energy and the calculated energy of the recoil nucleus assuming two-body kinematics. The emission spectrum becomes more energetic with increasing energy of the incident neutron.